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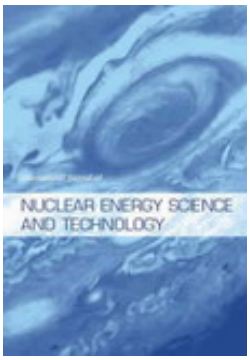

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Abstract: Innovative Compact Molten Salt Reactor (ICMSR) is a nuclear reactor designed to utilise thorium as main fertile fuel to achieve sustainable fuel resources. ICMSR has the inherent safety required as Advanced Reactor. This reactor uses liquid fuel salt. The fuel contains NaF-ThF_4-UF_4 (75-19.4-5.6)% mole of fuel salt with 19.75% uranium-235 enrichment. ICMSR uses graphite as moderator, Hastelloy-N as reactor vessel and NaF-KF (50-50)% mole as intermediate coolant. Fission yields produced by ICMSR include Xe and Kr. These isotopes need to be removed from the reactor because it reduces fuel utilisation. This paper describes the effect of Xe and Kr isotopes extraction to the criticality of ICMSR. Calculation is done by using MCNP6. The results show that the extraction of Xe and Kr on ICMSR increased the criticality and produced 2.661×10^8 Ci actinide fission products.

Keywords: ICMSR fuel; thorium sustainable fuel; Xe and Kr extraction; criticality; fission yield.

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1 Introduction

Nuclear energy is predicted to be the most important alternative energy source for the next few decades. Some of the reasons are: the world's growing energy needs, limited availability of conventional energy sources, nuclear energy system has been proven from technological and economic point of views.

Most commercial nuclear power plants currently use a light water reactor (LWR). LWR has disadvantages: new fuel must be added every 1–3 years due to low fissile breeding by uranium-238, not so high coolant operation temperature limits thermal efficiency to <35%, high pressure coolant, danger of steam explosion, high core radioactivity, high level waste disposal problem, high core afterheat during accidents, radiation damage limits to fuel and clad life, potential hydrogen generation during accidents, xenon-135 poisoning requires high excess core reactivity and careful power control (Dolan, 2017).

In 2002, innovations to obtain promising and more advanced nuclear energy in the needs and development of reactors were described in the Generation IV International Forum (GIF) (GIF, 2017). Referring to GIF, LWR does not qualify as a future reactor, which requires: more effective and efficient use of fuel for long-term energy availability worldwide, minimise the amount of waste generated, the probability of damage to the reactor core is very low, good passive safety system (GIF, 2015).

Among the many types of reactors, Molten Salt Reactor is one of six types of reactor that are included in the Advanced Reactor Category by GIF (World Nuclear Association, 2019). One of the most important things for advanced reactors is the design of the reactor model which is compact and capable to utilise fuel resources more efficient is increasing attention in design and deployment of next generation reactors. Innovative Compact Molten Salt Reactor (ICMSR) was proposed to solve all of these problems. ICMSR is molten salt fuel reactor where the fuel is also the coolant. ICMSR was chosen to optimise the use of thorium as a fertile fuel. So that the availability issue of uranium-235 can be solved because thorium is available in abundance. Thorium is 3 to 4 times more abundant than uranium in nature (GIF Expert Group, 2010). Thorium-based fuels also have the advantage of having inherent non-proliferation safeguards and less nuclear waste (Crossland, 2012; Du Toit and Chirayath, 2015; Lung and Gremm, 1998). The advantage of this reactor compared to solid fuel reactors is its ability to continuously remove fission products and add fissile or fertile elements.

During ICMSR operation, fission products are generated in the reactor. Some of them are neutron poisons, such as Xe and Kr. These isotopes can reduce the reactor criticality. So it is necessary to remove these isotopes from the reactor. Calculation of the effect of Xe and Kr extraction for Th-U sustainable ICMSR fuel was carried out using MCNP6 with ENDF/B-VII.I nuclear data. This computer code has been used for various types of

reactors that have complex geometry. Some examples of using MCNP are for calculations: reactor design, nuclear criticality safety and nuclear safeguard (Werner et al., 2017). This code was chosen because the benchmarking analysis showed a relatively small difference between similar computer programs and experimental data for commercial reactors (Jaradat and Alajo, 2017).

2 Fundamental theory and method

2.1 Innovative Compact Molten Salt Reactor (ICMSR)

Innovative Compact Molten Salt Reactor (ICMSR) uses liquid fuel salt, which is particularly suitable for utilising Th-U fuel cycle. The ICMSR fuel contains NaF-ThF_4-UF_4 (75-19.4-5.6)% mole of fuel salt with 19.75% uranium-235 enrichment. Uranium enrichment of less than 20% is intended to avoid attractive special grade nuclear materials (IAEA, 2001). ICMSR fuel salt does not need to be fabricated, so it is economically profitable. This fuel circulates through the core, pipes, pumps and heat exchangers. ICMSR is a newly proposed MSR adopting graphite as moderator, Hastelloy-N as reactor vessel and NaF-KF (50-50)% mole as intermediate coolant. ICMSR does not use control rods. Safety system is carried out based on the principle of gravity. In an emergency, the safety valve will melt so that there is a transfer of fuel from the core to the drain tank. So the reactor can be shutdown. Table 1 show the input parameters of ICMSR.

There are three nuclear materials in the ICMSR fuel: uranium-235, uranium-238, and thorium-232. Of the three isotopes, only uranium-235 can undergo a direct fission reaction. In general, the fission reaction can be written as:

$$ {}_0^1n + {}_Z^AX \rightarrow Y1 + Y2 + (2 \sim 3)n + energy(MeV) \tag{1} $$

In this case, Y1 and Y2 are fission product nuclides. Fission reactions can produce 2 to 3 neutrons, so a fission reaction can occur. Fission reactions also produce high energy, which is an average of 200 MeV (DOE, 1993).

Uranium-238 and thorium-232 are fertile materials. Fertile materials are materials that can undergo transmutation to become fissile materials. Both can produce other fissionable isotopes when exposed by neutrons. In the thorium fuel cycle, fertile Th-232 is converted to U-233. Reactors that are specifically designed to produce fissionable fuel are called breeder reactors (Lewis, 2008). U-233 is an isotope that does not exist in nature, so to maintain the initial fission chain reaction in the core is to use LEU.

ICMSR fuel containing fissile and fertile material can generate heat which will be converted to electrical energy by the turbine system. Figures 1 and 2 show the overall geometry of ICMSR. The output power of turbine system is 75 MWe, whereas the reactor thermal power is 187.5 MWth (thermodynamic efficiency = 40%).

ICMSR operates on the thermal spectrum so it requires a moderator to slow down the fast neutrons from fission into thermal neutrons. Possible moderators for thermal neutron spectrum Molten Salt Reactor have been investigated in ORNL, and graphite was selected as the most appropriate moderator (Lundy et al., 1960). The advantages of graphite are low neutron absorption cross-section, stable in radiation and high temperature environment, good mechanical strength at high temperature, and high thermal conductivity (Nightingale, 1966). Graphite also has a high neutron moderator

ratio and low thermal expansion (He et al., 2019). From an economic point of view, high-purity graphite is also the right choice because it can be easily found in the market at low prices (Atkinson et al., 2018). ICMSR thermal spectrum with graphite moderator provides better control and reduces initial fissile inventory. In addition to using graphite as a moderator, graphite is placed outside the core in reactor vessels to absorb neutrons and protect against neutron exposure.

Hastelloy-N is used in ICMSR reactor vessels, pipes, heat exchangers, and all metal components in contact with fluoride salts. Hastelloy-N is still considered a good material because it is most resistant to corrosion when interacting with reactor fuel at high temperature of 900K (Lundy et al., 1960).

Fluoride is used as molten salt because it is stable so it eliminates the risk of releasing radioactive material out of the reactor building. Fluoride can also lock volatile compounds or compounds that easily evaporate when under atmospheric pressure as iodine and cesium so that they cannot escape into the environment (Hargraves and Moir, 2010). Various studies have shown that fluoride fuel with graphite moderator has a negative coefficient of temperature reactivity (Zhang et al., 2009). Fluoride salt which is also used as coolant offer a higher neutron scattering ratio compared to conventional coolant. This higher scattering ratio increases the rate of conversion from fertile thorium-232 to fissile uranium-233. Actinium fluoride dissolved in the fuel salt, circulate between a graphite moderating region and a heat exchanger. When the fuel salt is in the moderation region, neutrons become sufficiently thermalised to sustain the fission reaction and generate heat. When leaving the moderation region, the fuel flows to the heat exchanger through the pipe, where the heat is transferred to the secondary side to be converted into electricity (Serp et al., 2014).

ICMSR has several important benefits, i.e. very low core damage frequency, the reactivity coefficient is negative in equilibrium, fuel reduction coefficient is negative in the equilibrium core condition, implement a totally passive safety system, the power feedback coefficient is negative, the excess reactivity is so low that the reactor becomes subcritical and the power drops. In ICMSR, a higher temperature rise will melt the fuel safety valve so that the fuel flows into the drain tank (inherent safety).

Table 1 Input parameter of ICMSR

Parameter	*Value (unit)*
Power	75 MWe; 187.5 MWth
Temperature (during operation)	900K
Fuel	$NaF\text{-}ThF_4\text{-}UF_4$
U-235 enrichment	19.75%
Core diameter; high	220 cm; 246 cm
Moderator	Graphite
Moderator diameter; high	28 cm; 150 cm
Intermediate coolant	NaF-KF
Vessel reactor	Hastelloy-N
Vessel diameter; high	400 cm; 10.51 m

Figure 1 Configuration of modular 75 MWe ICMSR system

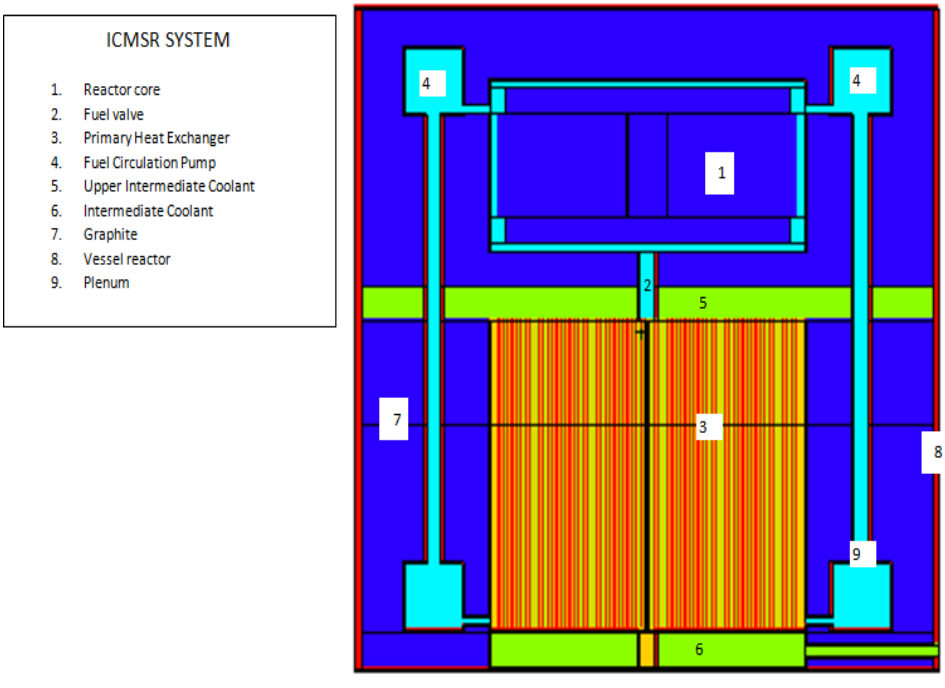
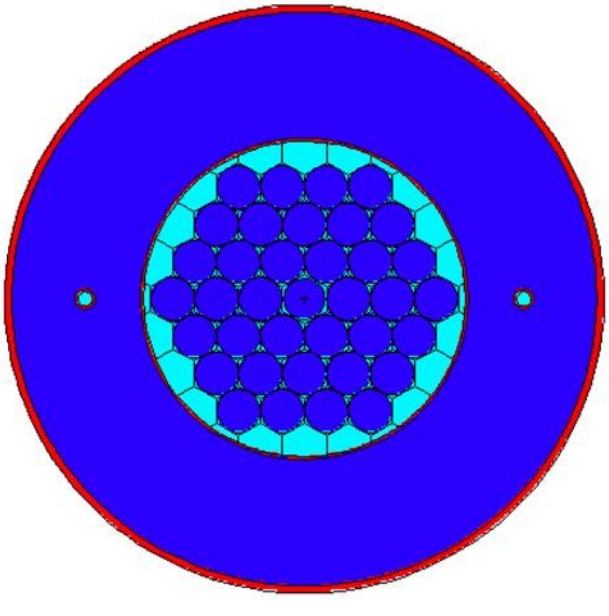


Figure 2 ICMSR geometry simulated by MCNP6 viewed from horizontal plane



ICMSR operation will generate fission products. Some of them have a high absorption cross section so that it affects the reactor criticality. The fission products that will be examined in this study are xenon and krypton. Xenon and krypton are noble gases. The noble and volatile gases have low solubility in molten salt operating temperature. These gases are easily removed by purging an inert gas into molten salt. However, this purging should not be done directly into molten salt fuel in reactor, because this purging will generate positive reactivity feedback. This purging should be done outside the reactor. The noble gases after extraction are flowed into the activated carbon bed for adsorption.

The solubility of noble gases in molten salts follows Henry's law, where pressure is proportional to the mole fraction (Benes and Konings, 2012):

$$p_i = k_H x_i \tag{2}$$

Engel et al. (1979) had reported that this purging method able to remove 80% of Xe-135 in MSRE. While Rosenthal et al. (1972) had shown that noble metal compound did not formed in molten salt fuel operating condition.

I-135 which is a xenon precursor can be removed from the reactor by the same method as xenon. While for samarium which is a non-noble gas fission product, the method is different from the noble gas. The non-gases fission products have varying solubility in molten salt fuels. The non-gases fission products also form fluoride compounds with different boiling points. The lower boiling point compounds tend to become volatile compound which can be treated with gas purging as explained before. The other compounds which have medium boiling points can be extracted with multistage distillation. The compounds which have relatively high boiling points can be extracted by metal-salt extraction method. This method is based on solubility difference of these compounds in liquid metal (i.e. Bi) and in molten salt fluoride. Same compound which have relatively low solubility can be extracted using electric precipitation method. All of these methods will be performed in a molten salt fuel processing system outside the reactor.

2.1 Calculation on MCNP

The Monte Carlo method simulates a large number of individual particle trajectories, and then averages the results to determine the desired quantities. Every question regarding the history of a neutron is decided by statistics. The Monte Carlo method is considered good and suitable for complex reactor geometries (Dolan, 2017). This method is used in MCNP software. MCNP is using Monte Carlo method and neutron transport. Monte Carlo for N-Particle Transport Version 6.0 (MCNP6) utilises nuclear continuous energy data ENDF/B-VII.I is released by Los Alamous National Laboratory (LANL). This software has calculation results that have been validated and used by many researchers for other examples MSR reactors (Jeong et al., 2016; Jaradat and Alajo, 2017).

One of the values that needs to be entered into MCNP6 is the density of fuel salt and coolant. The calculation of the density of the molten elements at the higher temperatures may be estimated from the equation (3) and Table 2 (Janz, 1988; Nasch and Steinemann, 1995):

$$\rho(t) = \rho_m - k\left(t - t_{\max}\right) \tag{3}$$

Table 2 Parameter values for molten element density

Formula	*Name*	t_m/°C	ρ_m/g cm^{-3}	k/g cm^{-3}°C^{-1}
NaF	Sodium Fluoride	996	1.948	0.000636
ThF_4	Thorium Fluoride	1110	6.058	0.000759
UF_4	Uranium (IV) Fluoride	1036	6.485	0.000992
KF	Potassium Fluoride	858	1.910	0.000651

The density value of this molten element will be used to find mixture density based on the mass fraction. The mixture density value will enter the MCNP6 material card and the ICMSR reactor geometry will be displayed in the Visual Editor (VisEd).

The criticality of a reactor can be expressed with a quantity called the effective multiplication factor which is symbolised by ke The k_{eff} number shows the ratio between the number of neutrons produced in the (i+1) generation and the number of neutrons produced by fission in the (i) generation. The number is systematically written as

$$k_{eff} = \frac{N_{i+1}}{N_i} \tag{4}$$

In a finite size system, some neutrons leak out of the system. The critical condition of the system must consider the neutron leakage. The effective multiplication factor can be written

$$k_{eff} = \frac{rate\,of\ neutron\ production}{rate\,of\ neutron\ absorption + neutron\ leakage} \tag{5}$$

In MCNP the definition of k_{eff} is written:

$$k_{eff} = \frac{\rho_a \int_V \int_0^\infty \int_E \int_\Omega \nu \sigma_f \Phi dV\, dt\, dE\, d\Omega}{\int_V \int_0^\infty \int_E \int_\Omega \nabla \cdot J\, dV\, dt\, dE\, d\Phi + \rho_a \int_V \int_0^\infty \int_E \int_\Omega \left(\sigma_c + \sigma_f + \sigma_m\right) \Phi dV\, dt\, dE\, d\Omega} \tag{6}$$

The requirement for criticality in a finite system without external neutron source is that $k_{eff} = 1$. $k_{eff} < 1$ means subcritical system. In a subcritical system, the number of neutrons will continue to decrease. $k_{eff} > 1$ means supercritical system. In a supercritical system, the production of neutrons is greater than the loss of neutrons, therefore the neutron population will tend to increase exponentially over time (DOE, 1993). In this study, the simulated neutrons in each cycle were set at 20,000 neutrons, with total of 250 cycles and the first 50 cycles were skipped.

Burn-up calculations are carried out at the reactor beginning of life conditions (plutonium free) and to obtain output values for the concentration of fission product. Among these nuclides, radioactive gases such as Xe and Kr have large neutron absorption cross sections, which impair the usefulness of neutrons during normal operation. In liquid fuels, these gases are not dissolved in molten salt (Compere et al., 1975; Kedl, 1972). Therefore, it can be extracted from molten salt and then stored for a sufficient decay period. The extraction of Xe and Kr is calculated to obtain a definite value which is expected to improve the safety system and the sustainability of ICMSR operations. For fission yields inventory calculation, BURN Card added which includes parameters TIME, POWER, PFRAC, MAT, OMIT, and more.

In general, the composition of a nuclide which is the result of a neutron reaction with the material in the reactor can be formulated as follows:

$$\frac{dN_i^z(t)}{dt} = \sum_{j\neq i} f_{j\to i}\lambda_j N_j^z(t) + \sum_{k\neq i} fact(t)\left\{g_{k\to i}C_k^z + \gamma_{k\to i}F_k^z + h_{k\to i}W_k^z\right\}N_k^z(t) - \left[\lambda_i + Fact(t)\left\{A_i^z + W_i^z\right\}\right]N_i^z(t) \quad (7)$$

Where:

i, j, k : depleted nuclide number

z : region number of burn up

N : burn up nuclide number density

λ, f : decay constant and branch ratio

g, γ, h: yield fraction of each transmutation

F : relative microscopic fission reaction rate

A : relative microscopic absorption reaction rate

C : relative microscopic capture reaction rate

W : relative microscopic (n, $2n$) reaction rate

fact : normalisation factor to convert relative reaction rates to absolute one

The meaning of equation (7) is nuclide i can be obtained by the decay process of nuclide j, neutron capture by nuclide k, fission reaction and (2,2n) reaction. At the same time, nuclide i is reduced by the process of self-decay, neutron absorption into other nuclides, and the (n, $2n$) reactions that converts itself to a lighter nuclide.

3 Result and discussion

Burn up calculations has been carried out for 40 months of operation with duration of 100 days. The reactor is operated at full power (187.5 MWth) without refuelling and changing the power. Running is done twice to see the effect of Xe and Kr extraction on criticality.

Figure 3 shows the accumulation of Kr-85 fission products. In general, the formation of Kr-85 from the fission process is linier to the fuel burn up. Kr-85 has a half-life of 10.73 years, Kr-85 will release two of β^- with an average energy of 47.5 keV and 251.4 keV, and also gamma rays with an energy of 514 keV. The daughter nuclide of Kr-85 decay is Rb-85. The total Kr-85 produced after 40 months of burn up is 8.695 x 10^4 Ci.

Figure 3 Kr-85 concentration for 40 months of ICMSR operation

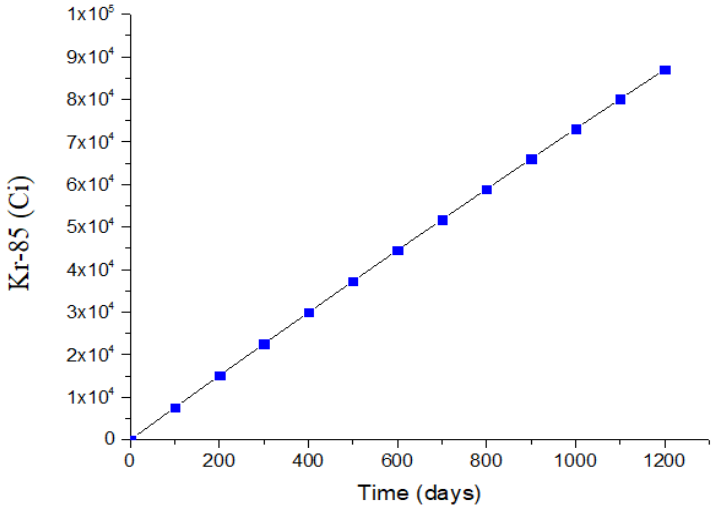
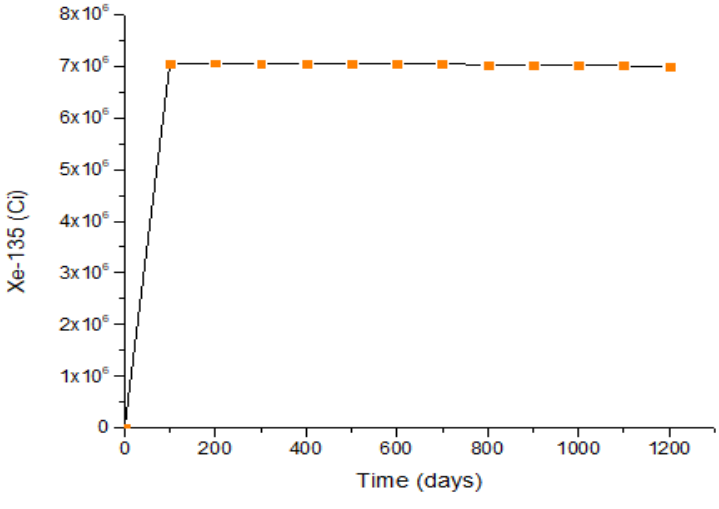


Figure 4 shows the accumulation of Xe-135 fission products. Xe-135 (9.1 hour half-life) which has the largest neutron capture cross section (2.6 million barn) of all known isotopes. The concentration of Xe-135 reaches saturation levels after passing through the 100th day. This is because the Xe-135 isotope produced is the same as the one that decays. The total Xe-135 produced after 40 months of burn up is 6.995×10^6 Ci. The father nuclide I-135 with a half-life of 6.61 hours contributes to an increase in the concentration of Xe-135. I-135 decays to Xe-135 which can affect reactivity and reduce energy production. In addition, another source of Xe-135 originates from the fission of U-235 and results from neutron absorption of Xe-135 within graphite. If Xe-135 does not undergo neutron capture, it can decay into Cs-135, a high-yield, long-lived isotope. These reasons show the importance of extracting Xe-135 from ICMSR.

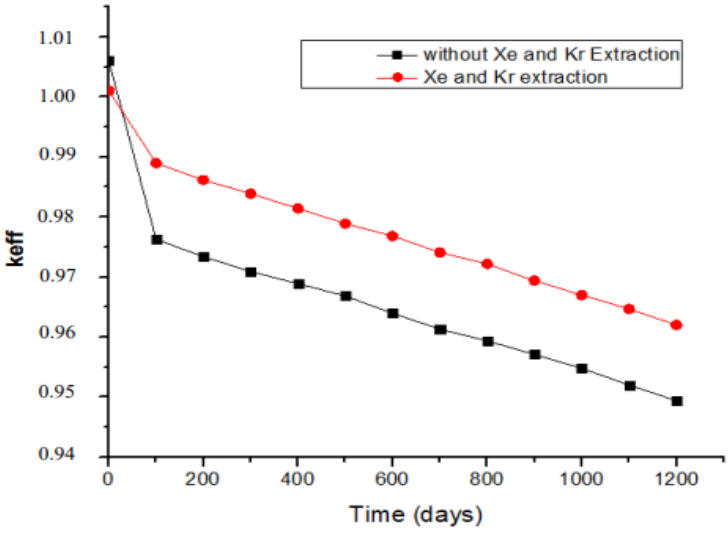
Figure 4 Xe-135 concentration for 40 months of ICMSR operation



Whereas, some other Xe and Kr isotopes have small absorption cross-sections and short half-lives so that they do not have a significant effect on reactor operation.

The effective multiplication factor continues to decrease gradually as the fissile material continues to be depleted of fuel salts during reactor operation. The presence of Xe and Kr in the reactor resulted in reduced effectiveness of fuel utilisation. It can be seen in Figure 5 that the k_{eff} value increases after extraction of Xe and Kr. In the first 100 days of operation of the reactor, the k_{eff} value for the extraction of Xe and Kr was closer to 1 so that the control of reactivity and safety was better. Therefore, removing Xe and Kr provides with significant neutron benefit and enables a longer core lifetime.

Figure 5 ICMSR criticality for 40 months



Xe and Kr isotopes that have been extracted from ICMSR should not be considered as nuclear waste material because they can be used for applications as recognised diagnostic-based reagents in nuclear medicine. The present paper revisits the use of Xe and Kr radioisotopes for radio-diagnostics but also for β^- radiotherapy of viral lungs diseases (Parker et al., 2012; Lull et al., 1983; Bailey and Roach, 2020; Degueldre et al., 2021).

Table 3 shows the main actinide composition in an ICMSR after extraction. Some actinides undergo transmutation, accompanied by neutron capture, negative beta decay, or alpha decay. The nuclide transmutation chain of ICMSR is shown at Figure 6. The actinide product formed after 40 months of burn up without the additional of fuel is 2.661×10^8 Ci.

Figure 6 Actinide transmutation chain of ICMSR fuel

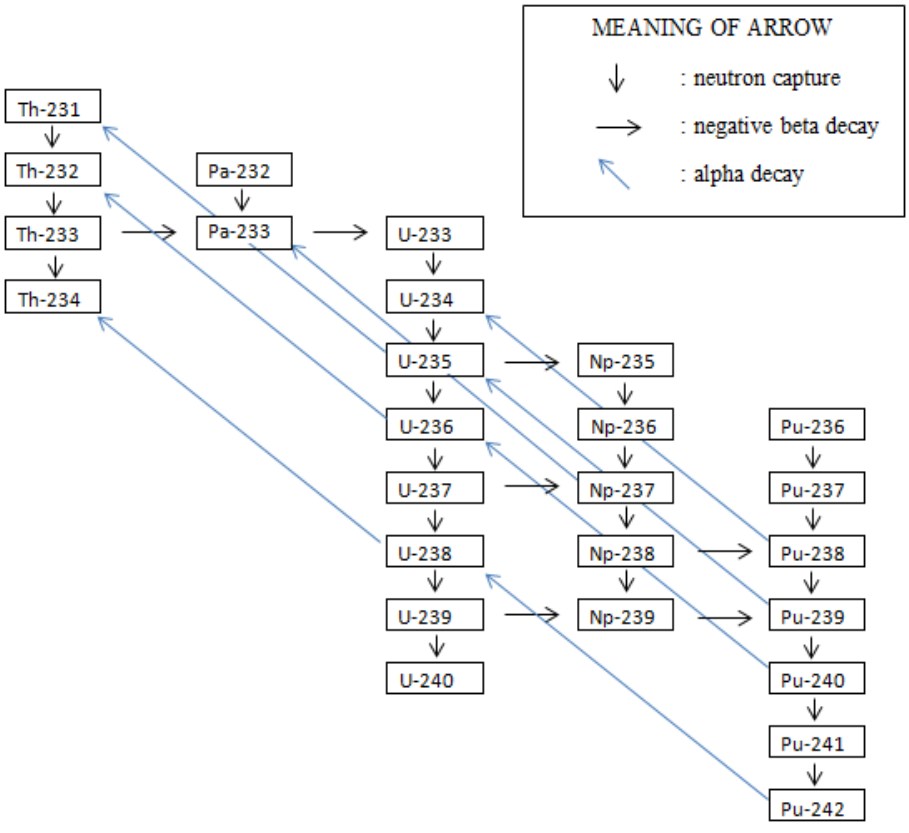


Table 3 Actinide composition of ICMSR at full power of 187.5 MWth

No.	*Isotope*	*Mass (g)*	*Activity (Ci)*
1	Th-231	2.968E-01	1.577E+05
2	Th-232	1.779E+07	1.951E+00
3	Th-233	2.712E+00	9.812E+07
4	Th-234	6.412E-05	1.484E+00
5	Pa-232	8.137E-02	3.495E+04
6	Pa-233	4.706E+03	9.765E+07
7	U-233	1.182E+05	1.139E+03
8	U-234	2.832E+03	1.760E+01
9	U-235	7.818E+05	1.690E+00
10	U-236	4.770E+04	3.085E+00
11	U-237	1.602E+01	1.307E+06
12	U-238	4.209E+06	1.415E+00
13	U-239	1.024E+00	3.431E+07
14	U-240	3.254E-15	3.013E-09

Table 3 Actinide composition of ICMSR at full power of 187.5 MWth (continued)

No.	*Isotope*	*Mass (g)*	*Activity (Ci)*
15	Np-235	6.769E-07	9.494E-04
16	Np-236	5.312E-04	6.996E-06
17	Np-237	1.022E+03	7.199E-01
18	Np-238	4.897E-01	1.269E+05
19	Np-239	1.479E+02	3.430E+07
20	Pu-236	3.848E-05	2.009E-02
21	Pu-237	1.911E-05	2.330E-01
22	Pu-238	6.684E+01	1.145E+03
23	Pu-239	3.622E+04	2.247E+03
24	Pu-240	3.275E+03	7.432E+02
25	Pu-241	1.177E+03	1.217E+05
26	Pu-242	6.034E+01	2.386E-01
	Totals	2.300E+07	2.661E+08

4 Conclusions

Burn up has been carried out on the ICMSR reactor to see the effect of Xe and Kr extraction on the operation of the reactor. The main attention is directed to isotopes having high absorption cross-sections and long half-lives. Extraction of Xe and Kr increased the criticality and produced 2.661×10^8 Ci actinide fission product.

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